

Force field study of dinitrogen tetroxide

K. RAMASWAMY AND T. SRIDHARAN

Department of Physics, Annamalai University, Annamalainagar, Tamilnadu

The force field study of Dinitrogen Tetroxide was carried out using parametric representation method. The parameter angles for the B_{1g} , B_{2u} and B_{3u} species were fixed using Isotopic frequency and were $\Phi = 26^\circ 44'$, $\Phi = 16^\circ 39'$ and $\Phi = 59^\circ 46'$ respectively. Strey's extremal condition on force constants was used in addition to the Isotopic frequencies to fix the best fit set of Eulerian angle parameters for the $3 \times 3 A_{1g}$ species. They are $\Psi = 48^\circ$, $\theta = 78^\circ 38'$ and $\Phi = 75^\circ 5'$. The force constants, mean amplitudes of vibration, Interaction Coriolis coupling constant etc., were calculated and compared with literature values.

1 INTRODUCTION

The study of a unique force field for polyatomic molecules by the method of parametric representation technique for 2×2 vibrational problems has attracted the attention of many workers (Torok 1967; Strey 1967; Jordanov *et. al* 1972; 1973; Redington *et. al* 1971, Isotani 1972, 1974; Ananthakrishnan *et. al* 1972). Recently an attempt was made to extend the parametric method to 3×3 vibrational problem (Ramaswamy *et. al* 1975) using Eulerian angles and with additional data like Coriolis coupling constant, isotopic shifts, mean vibrational amplitudes and rotational distortion constants. The present paper deals with the study of Dinitrogen Tetroxide.

The problem of the structure and the nature of bonding in N_2O_4 has been of considerable interest since N_2O_4 is expected to form a planar structure through the formation of a weak N-N bond. The planarity of the molecule involves the barrier to internal rotation. The theoretical considerations responsible for planarity has been made by several discussions (Bent 1963, Coulson *et. al* 1957, Smith *et. al* 1956, Green *et. al* 1961, Brown *et. al* 1961, Skancke *et. al* 1973). It is of interest to calculate the inertia defect of this molecule using a unique set of force constants which can be evaluated by the parametric representation technique using isotopic frequencies.

2. THEORETICAL CONSIDERATIONS

Planar N_2O_4 exhibits D_{2h} symmetry, for this symmetry, group theory shows that the twelve fundamental vibrations are divided among the irreducible representations in the following manner

$$3A_g + A_u + 2B_{1g} + B_{1u} + B_{2g} + 2B_{2u} + 2B_{3u}$$

The procedure to solve the inverse vibrational problem for the molecules like N_2O_4 is described below.

The force constant matrix F is given by

$$F = L_0^{-1} A \Lambda \tilde{A} L_0^{-1} \quad \dots\dots (1)$$

where L_0 is the genuine matrix of transformation between the symmetry coordinates and normal coordinates and also satisfies the normalising condition

$L_0 \tilde{L}_0 = G$. A is the properly chosen orthogonal parameter matrix and $\Lambda = 4\pi^2 c^2 \nu^2$ with the usual notations.

The proper orthogonal matrix A is of the order $n \times n$ for n^{th} order vibrational problem and needs $\frac{n(n-1)}{2}$ parameters. For 2×2 cases A is usually chosen as

$$A = \begin{vmatrix} C_\phi & S_\phi \\ -S_\phi & C_\phi \end{vmatrix} \quad (2)$$

For 3×3 cases A can be represented through the three Eulerian angles Ψ , θ and ϕ as

$$A = \begin{vmatrix} C_\Psi & S_\Psi & 0 \\ -S_\Psi & C_\Psi & 0 \\ 0 & 0 & 1 \end{vmatrix} \begin{vmatrix} 1 & 0 & 0 \\ 0 & C_\theta & S_\theta \\ 0 & -S_\theta & C_\theta \end{vmatrix} \begin{vmatrix} C_\phi & S_\phi & 0 \\ -S_\phi & C_\phi & 0 \\ 0 & 0 & 1 \end{vmatrix} \quad (3)$$

where C stands for *cosine* and S for *sine* of the respective angles. Following the method of Jordarov and Nikolova, (1972) the relation between the isotopic shifts and the parameter matrix is given by

$$\frac{K}{\Lambda} = \tilde{A} B A \quad \dots\dots (4)$$

where K is defined as the *kinematic* term in the differential form of Wilsons (1958) secular equation and $B = L_0^{-1} \Delta G \tilde{L}_0^{-1}$. Here ΔG is the change in the G matrix between the parent and the isotopic molecules concerned.

For 2×2 species the single parameter Φ can be directly obtained using the relation given by Ananthakrishnan & Aruldass (1972) as

$$\tan \phi_1 = \frac{B_{11} \pm \left[B_{11}^2 - \left(B_{11} - \frac{K_{11}}{\Lambda_1} \right) \left(B_{11} - \frac{K_{11}}{\Lambda_1} \right) \right]^{\frac{1}{2}}}{\left(B_{11} - \frac{K_{11}}{\Lambda_1} \right)} \quad (5)$$

and

$$\tan \phi_2 = \frac{-B_{11} \pm \left[B_{11}^2 - \left(B_{11} - \frac{K_{11}}{\Lambda_1} \right) \left(B_{11} - \frac{K_{11}}{\Lambda_1} \right) \right]^{\frac{1}{2}}}{\left(B_{11} - \frac{K_{11}}{\Lambda_1} \right)} \quad (6)$$

Here B_{ij} 's are the elements of B matrix and $\frac{K_{11}}{\Delta_1} + \frac{K_{11}}{\Delta_2} = B_{11} + B_{11}$.

The proper Φ which reproduces all the molecular constants has been selected as the best fit parameter. For the 3×3 species, the expansion of eq. (4) in terms of the B and A matrix elements leads to the following set of relations

$$\frac{K_{11}}{\Lambda_1} = p \frac{C^2}{\phi} + q \frac{S^2}{\phi} + 2r \frac{S}{\phi} \frac{C}{\phi} \quad (7)$$

$$\frac{K_{22}}{\Lambda_2} = p \frac{S^2}{\phi} + q \frac{C^2}{\phi} - 2r \frac{S}{\phi} \frac{C}{\phi} \quad (8)$$

and

$$\frac{K_{33}}{\Lambda_3} = [B_{11} \frac{S^2}{\psi} + B_{22} \frac{C^2}{\psi} + 2B_{12} \frac{S}{\psi} \frac{C}{\psi}] \frac{S^2}{\theta} + B_{33} \frac{C^2}{\theta} + 2[B_{13} \frac{S}{\psi} + B_{23} \frac{C}{\psi}] \frac{S}{\theta} \frac{C}{\theta} \quad (9)$$

where

$$p = [B_{11} \frac{C^2}{\psi} + B_{22} \frac{S^2}{\psi} - 2B_{12} \frac{S}{\psi} \frac{C}{\psi}]$$

$$q = [B_{11} \frac{S^2}{\psi} + B_{22} \frac{C^2}{\psi} + 2B_{12} \frac{S}{\psi} \frac{C}{\psi}] \frac{C^2}{\theta} + B_{33} \frac{S^2}{\theta} - 2[B_{13} \frac{S}{\psi} + B_{23} \frac{C}{\psi}] \frac{S}{\theta} \frac{C}{\theta}$$

and

$$r = [B_{12} (S_{\Psi}^2 - C_{\Psi}^2) + (B_{22} - B_{11}) S_{\Psi} C_{\Psi}] C_{\theta} + [B_{13} C_{\Psi} - B_{23} S_{\Psi}]$$

The sum rule is $\sum \frac{K_{ii}}{\Lambda_{ii}} = \sum B_{ii}$.

Eq. (9) involves Ψ and θ only. For known values of Λ ; Ψ can be varied in the interval of -90° to 90° to get two sets of θ values for every value of Ψ . This set of Ψ and θ on substitution in eq. (7) and or (8) yield different values of Φ . Thus for a given value of Ψ one obtains 2 values of θ and 4 values of Φ . They can be represented as (Ψ, θ_1, Φ_1) ; (Ψ, θ_1, Φ_2) ; (Ψ, θ_2, Φ_3) and (Ψ, θ_2, Φ_4) .

Out of these four sets the proper combination of (Ψ, θ, Φ) can be chosen using additional data like mean amplitudes of vibration, Coriolis coupling constants and rotational distortion constants whenever is available. For N_2O_4 , only electron diffraction results are available. Hence attempt was made to fix the A matrix using the mean vibrational amplitudes of $N-N$ and $N=O$ bonds, as Coriolis coupling constants and rotational distortion constants are not available.

For every value of Ψ in the interval -90° to 90° , the mean vibrational amplitude of $N-N$ and $N=O$ were calculated using Cyvin's (1968) relation $\Sigma = L \Delta \bar{L}$.

where L is defined as $L = L_0 A$

Graphs are drawn between Ψ and mean amplitudes of vibration for the four sets of (Ψ, θ, Φ) 's. That value of Ψ which corresponds to the observed mean amplitude of vibration is chosen and hence the proper combination of (Ψ, θ, Φ) is found out.

3. RESULTS AND DISCUSSION

The Infrared studies of N_2O_4 have been reported in gaseous condensed phases by several authors (Begun *et al.* 1960, Snyder *et al.* 1957, Hisatsune 1959, Wiener *et al.* 1957, Sutherland 1933, Strony *et al.* 1932, Schaffert 1933, Cassie *et al.* 1933, Harris 1934). Begun & Fletcher (1960) have the most complete assignment of the N_2O_4 spectrum based on isotopic shift data table 4. However, the torsional mode frequencies which can provide information on the barrier to internal rotation have not been observed. Bibart *et al.* (1974) identified the sequence bands, which enable torsional frequencies to be determined. Together with previous Raman results and a third law entropy

Table 1. Vibrational frequencies (cm^{-1}) and molecular parameters for N_2O_4

Species	Vibrational frequencies in cm^{-1}		Molecular parameters
	$^{14}\text{N}_2\text{O}_4$	$^{15}\text{N}_2\text{O}_4$	
A_{1g}	1379.6	1360.2	$D_{\text{N-N}} = 1.782 \text{ \AA}$ $d_{\text{N=O}} = 1.190 \text{ \AA}$ $\angle \text{O-N-O} = 135^\circ 24'$
	808	797	
	256	264	
B_{1g}	1712	1672	
	482	478	
B_{2u}	1748	1707	
	381	377	
B_{3u}	1262	1251	
	750	739	

calculation, they determined the remaining unknown vibrational frequency, and characterised the potential which restricts the internal rotation in N_2O_4 . From the potential function a picture of the bonding in N_2O_4 is presented. Finally they suggested that in addition to the N—N bond, weak σ bonds form between the cis oxygens which stabilize the eclipsed oxygen configurations and tend to lock N_2O_4 molecule into a planar configuration. McClelland, Gunderson & Hedberg (1972) reinvestigated the structure of N_2O_4 by gas electron diffraction and suggested that the very long N—N bond and its large amplitude of vibration implies a weak link and are in accord with the low dissociation energy. Tevault & Andrews (1974) also studied about the Raman spectrum of N_2O_4 recently. Hisatsune *et al.* (1960) calculated the Urey-Bradley potential constants of N_2O_4 . Green's function analysis of N_2O_4 was done by Ramaswamy & Mohan (1971).

The graphs for Ψ for various values of calculated mean amplitudes of vibration are shown in figure (1). It is seen from figure (1) that the calculated mean amplitudes show a discontinuity in the region -30 to 45° due to the fact that the value of discriminant in eqs. (5) and (6) became imaginary. It is also seen from the graph that the observed N—N mean amplitude is higher than the calculated values and also the calculated mean amplitudes lie within the limiting values. The maximum value which is permissible from the graph for N—N is 0.0765 and minimum is 0.0716. Since these calculated values lie within the values of 0.0816 ± 0.0178 it is unfortunately

not possible to unambiguously fix (Ψ, θ, Φ) using bonded N-N mean amplitudes.

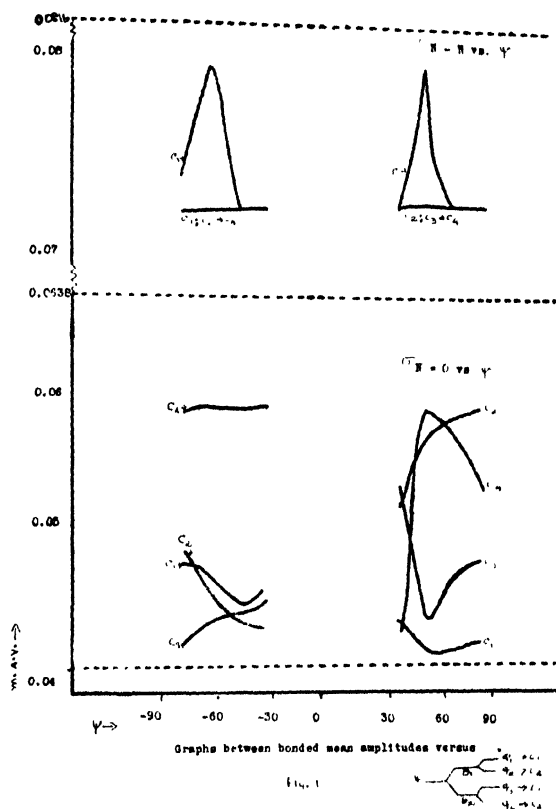


Fig. 1

Similarly in the case of σ_{N-O} bonded mean amplitudes all the calculated values are higher than the observed value (maximum calculated value = 0.0598 and minimum = 0.0412. Observed = 0.0381 ± 0.0019). Hence it is not also possible to fix uniquely using bonded $\sigma_{N=O}$ mean amplitudes.

In order to fix the Ψ, θ and Φ parameters for the A species, Strey's (1967) extremal properties of force constant was used. Since Dinitrogen Tetroxide is a non-hydride molecule, the condition of bending force constant is a minimum, is applied. The bending force constants for all the four combination of A matrices are calculated. By this means sets of A matrices are obtained, out of these the proper A matrix is selected, which reproduced all the molecular constants which are in agreement with those of earlier workers. The angle parameters for the best fit A matrix are $\Psi = 48^\circ$, $\theta = 76^\circ 38'$ and

$\Phi = 75^\circ 5'$. The symmetrised force constant calculated using the above set compares well with the values of Ramaswamy & Mohan (1971).

For the $2 \times 2 B_{1g}$ species, the use of $\frac{K_{55}}{\Lambda_5}$ isotopic shift in eq. (5) gave two solutions for Φ and are $\Phi = -31^\circ 40'$ and $25^\circ 1'$. The use of $\frac{K_{66}}{\Lambda_6}$ in eq. (6) gave two more solutions for Φ and are $\Phi = -35^\circ 6'$ and $28^\circ 26'$. The discrepancy in the angle parameters calculated is due to the slight disagreement in the sum rule $\Sigma \frac{K_{ii}}{\Lambda_{ii}} = \Sigma B_{ii}$. Hence the average values of $\Phi = -33^\circ 23'$ and $26^\circ 44'$ are considered to fix the force fields. Of these two angles, $\Phi = 26^\circ 44'$ reproduced the force constants which are in agreement with those of Ramaswamy *et. al.* (1971) and hence taken as the best fit angle parameter for the B_{1g} species.

In the case of $2 \times 2 B_{2u}$ the use of $\frac{K_{99}}{\Lambda_9}$ isotopic shift in eq. (5) gives only one angle parameter viz, $\Phi = -4^\circ 25'$. Since the discriminant became imaginary. The use of $\frac{K_{1010}}{\Lambda_{10}}$ isotopic shift in eq. (6) gave two more angles and are $\Phi = -46^\circ 41'$ and $37^\circ 49'$. The wide discrepancy in the solutions are attributed to the disagreement in the sum rule $\Sigma B_{ii} = \frac{\Sigma K_{ii}}{\Lambda_{ii}}$. And hence the average values of $\Phi = -25^\circ 33'$ and $16^\circ 42'$ are considered and found that the value of $\Phi = 16^\circ 42'$ reproduced the diagonal force constants in reasonable agreement with the literature values.

Similarly for the $2 \times 2 B_{3u}$ species the isotopic shift $\frac{K_{11,11}}{\Lambda_{11}}$ reproduced the angles $\Phi = 60^\circ 42'$ and $\Phi_2 = -45^\circ 58'$. The use of $\frac{K_{12,12}}{\Lambda_{12}}$ gave two more angles and are $\Phi = 58^\circ 51'$ and $-44^\circ 7'$. The average values of $\Phi = 59^\circ 46'$ and $-45^\circ 2'$ are considered and out of these the best fit angle parameter chosen is $\Phi = 59^\circ 46'$.

The symmetrised force constants calculated in the present study compare well with those of Ramaswamy *et. al.* (1971) and are shown in table 2. Ladd & Orville-Thomas (1966) have proposed an empirical relationship between

Table 2. Symmetrised force constants (m.dyn/Å) for N_2O_4

Species and Angle parameters	F elements	Present work	Ramaswamy Mohan (1971)
$A_{1g} :$			
$\Psi = 48^\circ$	F_{11}	1 3017*	1 3587
$\theta = 76^\circ 38'$	F_{22}	12.7855	12.3639
$\varphi = 75^\circ 5'$	F_{33}	1.0656	0.7671
	F_{12}	1 4619	-1.1022
	F_{13}	-0.0666	-0.1331
	F_{23}	-0.4541	0.9735
$B_{1g} :$			
$\varphi = 26^\circ 44'$	F_{44}	10.0998	9.6069
	F_{55}	0.9491	0.6607
	F_{45}	0 7181	0 5000
$B_{2u} :$			
$\varphi = 16^\circ 19'$	F_{66}	10.8572	10 1514
	F_{77}	2 4777	1 2459
	F_{67}	2.6452	0 8103
$B_{3u} :$			
$\varphi = 59^\circ 46'$	F_{88}	11.9438	11 9535
	F_{99}	1.0595	0 7281
	F_{89}	0.6422	0.7720

*This number of significant figures are retained to secure internal consistency in calculations.

the N—O bond length and the corresponding force constant. Using their relationship the value obtained is 12 3262 m.dyn/Å for the N=O stretching force constant agrees well with the calculated value of 11.6461 m.dyn/Å. The mean vibrational amplitudes are calculated and compared with those of McClelland *et al's* electron diffraction results. It is found that the non-bonded mean amplitudes agrees quite well table 3. The potential energy distribution (Oka & Morino 1965) for all the best fit values were calculated and were shown in table 4. The interaction coriolis coupling constants were calculated and presented in table 5. Since experimental data is not available no direct comparison is possible. The calculated interaction Coriolis coupling constants obeys the sum rules given by Kuchitsu, Oka & Morino (1965). The inertia defect defect for this molecule is calculated using the torsional mode

frequency as $\nu_4 = 79$ given by Bibart *et al* (1974). This value of inertia defect $\Delta = -0.0744$ is not comparable with the ones reported by Jayapandian (1976) as he has used $\nu_4 = 160 \text{ cm}^{-1}$. The rotational distortion constant parameters (τ_{xxxx}) are also calculated presented in table 5.

Table 3. Calculated mean amplitudes of vibration (Å) for N_2O_4

	Present study	Ref. McClelland et. al. 1972
$\sigma_{\text{N-N}}$	0.0716	0.0816 ± 0.0178
$\sigma_{\text{N=O}}$	0.0421	0.0381 ± 0.0019
$\sigma_{01 \cdots 02}$	0.0486	0.0493 ± 0.0040
$\sigma_{01 \cdots \text{N2}}$	0.0747	0.0729 ± 0.0061
$\sigma_{01 \cdots 03}$	0.0984	0.0970 ± 0.0167
$01 \cdots 04$	0.0730	0.0730 ± 0.0114

Table 4. Potential energy distribution matrices for N_2O_4

A_g species	0.885671	0.065384	0.196479
	0.125875	0.894739	0.140832
	0.218740	0.137623	0.795199
B_{1g} species	1.080260	0.099221	
	0.000185	0.972262	
B_{2u} species	1.090705	0.112378	
	0.025389	1.077362	
B_{3u} species	1.056824	0.001247	
	0.079168	1.026980	

Table 5. Calculated interaction Coriolis coupling constants, Rotational distortion parameters (KHz) and Inertia defect for N_2O_4

$\xi_{4,11}^a = -0.7872$	$\xi_{1,8}^b = -0.0741$	$\xi_{1,5}^c = 0.1434$
$\xi_{4,12}^a = 0.6180$	$\xi_{2,8}^b = -0.6709$	$\xi_{2,5}^c = 0.6159$
$\xi_{6,8}^a = 0.9646$	$\xi_{3,8}^b = -0.7232$	$\xi_{3,5}^c = 0.7755$
$\xi_{6,8}^a = 0.2638$	$\xi_{4,10}^b = 0.3597$	$\xi_{1,6}^c = 0.7166$
$\xi_{7,9}^a = -0.9331$	$\xi_{4,10}^b = -0.9330$	$\xi_{2,6}^c = -0.6027$
$\xi_{7,10}^a = -0.3597$	$\xi_{7,11}^b = 0.6262$	$\xi_{3,6}^c = 0.3506$
	$\xi_{7,12}^b = 0.8132$	$\xi_{8,11}^c = 0.3011$
		$\xi_{10,11}^c = 0.9597$
$-h^4 \tau_{xxxx} = 1.8334$	$\Delta = -0.0744$	$\xi_{9,12}^c = 0.9821$
$-h^4 \tau_{yyvv} = 12.1083$		$\xi_{10,12}^c = -0.2841$
$-h^4 \tau_{zzzz} = 1.6557$		
$-h^4 \tau_{xyxy} = 4.0337$		

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